

Coherent spin dynamics of nanomolecules and magnetic nanoclusters

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Abstract

Spin dynamics of nanomolecules and nanoclusters are analyzed. The nanosizes of these objects make it possible to consider them as single-domain magnets with a large total spin, where the motion of the spins of all atoms, composing a nanocluster, occurs in a coherent way. Another meaning of coherence in spin dynamics is the coherent spin motion of several nanomolecules or nanoclusters. Different approaches for treating spin dynamics are compared and the main mechanisms influencing the spin motion are studied. Spin dynamics of separate magnetic nanomolecules and nanoclusters are investigated, as well as the spin dynamics of the ensembles of these nano-objects.

1 Introduction

Magnetic nanomolecules and nanoclusters enjoy many similar properties because of which the dynamics of their magnetization can be described by the same type of equations. This is why, we consider both these nano-objects together. Of course, there is difference in their structure and parameters which we shall take into account and characterize them by the appropriate models. The detailed description of general physical properties and applications of different magnetic nanoparticles can be found in review articles [1-9]. Here we briefly mention those of the properties and parameters that will be necessary for the following consideration.

It is worth stressing that there exist two types of magnetic nanoparticles. One large class consists of nanoclusters and nanomolecules, whose magnetic moments are formed by electron spins. Another type includes nanomolecules that possess magnetic moments solely due to polarized proton spins. Examples are propanediol $C_3H_8O_2$, butanol C_4H_9OH , and ammonia NH_3 . In such nanomolecules, there is no any other magnetic moment except that caused by polarized protons. So, here the proton magnetic moment is not a contribution, but the main object.

The magnetic moment of an atom is composed of electron and proton moments, with the electron magnetic moment $\mu_e = -g_e\mu_B S = \hbar\gamma_e S = -\mu_B$ and the proton magnetic moment

$\mu_p = g_p \mu_N S = \hbar \gamma_p S$, where $g_e = 2$ and $g_p = 5.586$ are the electron and proton Landé factors, $\mu_B = |e|\hbar/2m_e$ and $\mu_N = |e|\hbar/2m_p$ are the Bohr and nuclear magnetons, γ_e and γ_p are the electron and proton gyromagnetic ratios. Since the proton mass is larger than that of an electron, $m_p/m_e \sim 10^3$, the proton magnetic moment is essentially smaller, $\mu_p/\mu_e \sim 10^{-3}$. The electron and proton radii are $r_e \sim 10^{-15}\text{cm}$ and $r_p \sim 10^{-13}\text{cm}$, respectively. An atom is called magnetic, when its total magnetic moment is nonzero. The total spin of a magnetic atom can be between $1/2$ and $S \sim 10$, hence, its magnetic moment can be of order $1\mu_B - 10\mu_B$. Atom radii are of order $r_A \sim 10^{-9} - 10^{-8}\text{cm}$. Examples of magnetic atoms are Fe (Iron), Co (Cobalt), Ni (Nickel), Gd (Gadolinium), and Cr (Chromium).

Magnetic nanomolecules are composed of many magnetic atoms and, as is clear from their name, are of the nanometer size. An important property of a magnetic nanomolecule is that its total magnetic moment can be treated as being due to an effective total spin. Generally, the molecule spin can be directed either up or down, with an energy barrier between these directions of order $E_A \sim 10 - 100$ K. At high temperatures, above the *blocking temperature* $T_B \sim 1 - 10$ K, a magnetic molecule behaves as a superparamagnetic particle, whose spin randomly oscillates between the up and down positions. While below the blocking temperature the spin is frozen in one of the directions.

Magnetic nanoclusters are also made of magnetic atoms that are assembled together in a random way. This distinguishes them from magnetic molecules, where atoms are strictly connected by chemical bonds. The sizes of nanoclusters can be in the range between 1 nm and 100 nm, containing about $100 - 10^5$ atoms. These values define the *coherence radius* R_{coh} , below which a nanocluster is in a single-domain state and can be treated as a large particle with an effective spin. A cluster, with a size larger than R_{coh} , separates into domains with opposite magnetizations. Similarly to magnetic molecules at low temperature, the magnetic moment of a nanocluster, below the blocking temperature $T_B \sim 10 - 100$ K, is frozen in one of two possible directions. The effective spin of a nanocluster is formed by electron spins and can be as large as $S \sim 100 - 10^5$.

The often considered nanoclusters are made of the magnetic atoms of Fe, Ni, and Co. They can be made of oxides, such as NiO, Fe_2O_3 , NiFe_2O_4 or alloys, such as $\text{Nd}_2\text{Fe}_{14}\text{B}$, $\text{Pr}_2\text{Fe}_{14}\text{B}$, $\text{Tb}_2\text{Fe}_{14}\text{B}$, DyFe_{14}B , $\text{Pr}_2\text{Co}_{14}\text{B}$, $\text{Sm}_1\text{Fe}_{11}\text{Ti}_1$, $\text{Sm}_1\text{Fe}_{10}\text{V}_2$, $\text{Sm}_2\text{Fe}_{17}\text{N}_{23}$, $\text{Sm}_2\text{Fe}_{17}\text{C}_{22}$, $\text{Sm}_2\text{Co}_{17}$, Sm_2Co_5 . To protect nanoclusters from oxidation, one coats them with graphene or noble metals, forming the double-component nanoclusters, such as Fe-Au, Co-Au, Co-Ag, Co-Cu, Co-Pt, Co-Pd, Ni-Au, Ni-Ag, Ni-Pd, and Mn^- -Au. The coating is done by means of chemical reactions or laser ablation techniques. The nanoclusters are produced by employing thermal decomposition, microemulsion reactions, and thermal spraying.

Magnetic nanoclusters and nanomolecules find numerous applications, among which we can mention magnetic chemistry, biomedical imaging, medical treatment, genetic engineering, waste cleaning, information storage, quantum computing, and creation of radiation devices. Since both nanomolecules and nanoclusters possess many common properties and can be considered as single particles with a large spin, we shall often talk on nanoclusters, implying that similar effects can be realized with both of them, molecules as well as clusters.

The use of these nano-objects requires the existence of two properties that contradict each other. From one side, to be able to keep memory, a cluster has to enjoy a stable state with its spin frozen in one direction. But from another side, in order to be able to manipulate the cluster magnetization, there should exist a way of suppressing the anisotropy. And it is necessary that the spin manipulation could be done sufficiently fast, so that the cluster

magnetization could be quickly reversed. Recall that thermal reversal is characterized by the Arrhenius law giving the longitudinal relaxation time $T_1 \sim \exp\{E_A/k_B T\}$, where E_A is the anisotropy energy, so that, at temperatures below the blocking temperature, the magnetization is frozen.

Magnetization reversal can be realized by different methods, by applying transverse constant or alternating magnetic fields and short magnetic field pulses [10]. To achieve fast reversal, one needs to find optimal values for the amplitude, frequency, and duration of such field pulses.

A very efficient method of achieving ultrafast magnetization reversal of magnetic nanoclusters has been suggested [11] by employing the acceleration effect caused by a resonator feedback field. The efficiency of this method is due to self-optimization of the spin motion producing the resonator field acting back on the spins. Historically, this effect was described by Purcell [12] and considered by Bloembergen and Pound [13] using classical phenomenological equations. Such equations are not sufficient for describing different regimes of spin motion. Microscopic theory of spin dynamics has been developed being applied to polarized proton spins of such molecules as propanediol $C_3H_8O_2$, butanol C_4H_9OH , and ammonia NH_3 (see review articles [4,14]) and to magnetic molecules [15-19].

The aim of the present paper is threefold. First, we concentrate on the spin dynamics of nanoclusters, comparing the peculiarity of their spin motion with that of proton and molecular spins. Second, we analyze the role of other effects, such as the Nyquist-noise triggering and Dicke correlation, studying their influence on the spin dynamics of nanoclusters. We show that these effects are negligible as compared to the Purcell effect. And, third, we compare different approaches to describing spin dynamics, demonstrating the advantage of using a microscopic approach based on quantum equations of motion.

2 Phenomenological classical equations

Dynamics of the magnetic moment \mathbf{M} of a magnetic particle is usually described by the classical equation

$$\frac{d\mathbf{M}}{dt} = -|\gamma_S|\mathbf{M} \times \mathbf{H}_{eff} + \mathbf{R} , \quad (1)$$

in which γ_S is the giromagnetic ratio of the particle with spin S and \mathbf{R} is a relaxation term. The effective magnetic field is given by the variational derivative $\mathbf{H}_{eff} = -\delta E/\delta\mathbf{M}$ of the particle energy E . The length of the magnetic moment is conserved, when the right-hand side of the equation $d\mathbf{M}^2/dt = 2\mathbf{M} \cdot \mathbf{R}$ is zero.

Choosing the relaxation term in the form

$$\mathbf{R} = -\frac{\alpha|\gamma_S|}{M} \mathbf{M} \times (\mathbf{M} \times \mathbf{H}_{eff}) , \quad (2)$$

one gets the Landau-Lifshitz equation, where α is a dissipation parameter and $M \equiv |\mathbf{M}|$. Under form (2), $|\mathbf{M}|$ is conserved. The equation was initially derived [20] for describing energy dissipation in the process of magnetic domain wall motion inside bulk ferromagnetic matter. Though it is often applied for treating the dynamics of ferromagnetic particles [21].

Taking the relaxation term as

$$\mathbf{R} = \frac{\alpha}{M} \mathbf{M} \times \frac{d\mathbf{M}}{dt} , \quad (3)$$

one comes to the Gilbert equation [22]. This equation, up to a renotation of parameters, is equivalent to the Landau-Lifshitz equation. Hence, it has the same region of applicability, though it is also used for describing the magnetization rotation of magnetic particles [10].

Another form of the relaxation term has been advanced by Bloch [23] as

$$\mathbf{R} = - \frac{M_x - M_x^*}{T_2} \mathbf{e}_x - \frac{M_y - M_y^*}{T_2} \mathbf{e}_y - \frac{M_z - M_z^*}{T_1} \mathbf{e}_z , \quad (4)$$

where \mathbf{M}^* is an equilibrium magnetization, \mathbf{e}_α are unit coordinate vectors, and the relaxation parameters are characterized by the longitudinal relaxation time T_1 and transverse relaxation time T_2 . The latter is also called the dephasing time. For an ensemble of N magnetic particles with a large average spin polarization

$$s \equiv \frac{1}{SN} \sum_{j=1}^N \langle S_j^z \rangle , \quad (5)$$

the transverse term has to be renormalized [16,24] as $1/\tilde{T}_2 = (1 - s^2)/T_2$.

The Landau-Lifshitz equation has a single dissipation parameter α and preserves spherical symmetry, thus, describing isotropic magnetization rotation. Because of these properties, it is appropriate for bulk macroscopic ferromagnetic matter with spherical magnetic symmetry. It may also be used for magnetic clusters, possessing this symmetry, which, however, is a rather rare case.

The Bloch equation has two relaxation parameters, T_1 and T_2 . Therefore it can describe more general situation of anisotropic relaxation, which is more realistic for treating nanoclusters in a medium or below the blocking temperature, when $T_2 \ll T_1$. The Bloch equations have been employed for considering the electron and nuclear spin motion in a strongly coherent regime [25-29] and for spin-polarized ^{129}Xe gas [30]. But these equations cannot describe the whole process of spin relaxation starting from an incoherent quantum stage, for which a microscopic approach is necessary [29,31,32]. The initial stage of spin relaxation is triggered by quantum spin fluctuations that can be identified with nonequilibrium spin waves [4,16,31-33].

3 Microscopic quantum approach

In a self-consistent quantum approach, we start with a microscopic spin Hamiltonian \hat{H} that is a functional of spin operators \mathbf{S} . The evolution equations are given by the Heisenberg equations of motion

$$i\hbar \frac{d\mathbf{S}}{dt} = [\mathbf{S}, \hat{H}] . \quad (6)$$

The advantage of using the quantum approach is in the following. First, it takes into account quantum effects that can be important for small clusters. Hence, it is more general. Second, at the initial stage of free spin relaxation, quantum spin fluctuations are of principal importance, being the triggering mechanism for starting the spin motion. Third, being based on an explicit spin Hamiltonian makes it possible to control the used approximations and to have well defined system parameters.

We assume that a magnetic cluster is inserted into an magnetic coil, of n turns and length l , of a resonant electric circuit characterized by resistance R , inductance L , and capacity C . The coil axis is taken along the axis x . Moving magnetic moments induce in the coil the electric current j described by the Kirchhoff equation

$$L \frac{dj}{dt} + Rj + \frac{1}{C} \int_0^t j dt = - \frac{d\Phi}{dt} + E_f , \quad (7)$$

in which the magnetic flux $\Phi = 4\pi n M_x / cl$ is formed by the mean transverse magnetization $M_x = \mu_0 \sum_{j=1}^N \langle S_j^x \rangle$, where $\mu_0 \equiv \hbar \gamma_S$. Here E_f is an additional electromotive force, if any. The resonator natural frequency and circuit damping, respectively, are

$$\omega = \frac{1}{\sqrt{LC}} , \quad \gamma = \frac{R}{2L} . \quad (8)$$

The coil current creates the magnetic field

$$H = \frac{4\pi n}{cl} j \quad (9)$$

that is the solution to the equation

$$\frac{dH}{dt} + 2\gamma H + \omega^2 \int_0^t H(t') dt' = -4\pi\eta \frac{dm_x}{dt} , \quad (10)$$

where $\eta \equiv V/V_{coil}$ is the filling factor and

$$m_x \equiv \frac{M_x}{V} = \frac{\mu_0}{V} \sum_{j=1}^N \langle S_j^x \rangle$$

is the transverse magnetization density. The external electromotive force is omitted. The field H is the feedback field, created by moving spins and acting back on them.

4 Dynamics of a single nanocluster

The typical Hamiltonian of a nanocluster is

$$\hat{H} = -\mu_0 \mathbf{B} \cdot \mathbf{S} - D(S^z)^2 + D_2(S^x)^2 + D_4 [(S^x)^2(S^y)^2 + (S^y)^2(S^z)^2 + (S^z)^2(S^x)^2] , \quad (11)$$

where the total magnetic field

$$\mathbf{B} = B_0 \mathbf{e}_z + B_1 \mathbf{e}_x + H \mathbf{e}_x \quad (12)$$

consists of an external constant field B_0 , weak transverse anisotropy field B_1 , and the feedback resonator field H . The anisotropy parameters D, D_2, D_4 are defined by the particular type of considered nanoclusters.

The main attention will be paid to the investigation of spin dynamics starting from a strongly nonequilibrium initial state, where the magnetization is directed opposite to the constant external magnetic field B_0 .

First, we study the influence of the thermal Nyquist noise of the coil in order to understand whether it can trigger the spin motion in a nanocluster. For the thermal-noise relaxation time, we find

$$t_T = \frac{4\gamma V_{coil}}{\hbar\gamma_S^2\omega} \tanh\left(\frac{\omega}{2\omega_T}\right), \quad (13)$$

where $\omega_T \equiv k_B T/\hbar$ is the thermal frequency defined by temperature T . At low temperatures, below the blocking temperature, say at $T = 1$ K, we have $\omega_T \sim 10^{12} \text{ s}^{-1}$. Then the thermal-noise relaxation time is

$$t_T \simeq \frac{2\gamma V_{coil}}{\hbar\gamma_S^2\omega_T} \quad \left(\frac{\omega}{\omega_T} \ll 1\right). \quad (14)$$

On the other side, for the reversal time, caused by the resonator feedback field, we have

$$t_{rev} \simeq \frac{V_{coil}}{\pi\hbar\gamma_S^2 S}. \quad (15)$$

The ratio of the latter to the thermal time (13) is $t_{rev}/t_T \sim \omega_T/2\pi\gamma S$. For the typical values $T = 1$ K, $\gamma \sim 10^{10} \text{ s}^{-1}$, and $S \sim 10^3$, this ratio is small: $t_{rev}/t_T \sim 10^{-2}$. Therefore the thermal Nyquist noise does not play any role in the spin dynamics of a nanocluster.

We have accomplished numerical solution of the evolution equations for nanocluster parameters typical of Fe, Ni, and Co nanoclusters. The Zeeman frequency is taken as $\omega_0 \equiv 2\mu_B B_0/\hbar \sim 10^{11} \text{ s}^{-1}$. For the feedback rate, we have $\gamma_0 \equiv \pi\eta\hbar\gamma_S^2 S/V_{coil} \sim 10^{10} \text{ s}^{-1}$. The typical anisotropy parameters satisfy the relations $D/(\hbar\gamma_0) \sim 10^{-3}$, $D_2/(\hbar\gamma_0) \sim 10^{-3}$, $D_4/(\hbar\gamma_0) \sim 10^{-10}$. At the initial time, the spin is assumed to be directed along the axis z . The resonator natural frequency is taken to be in resonance with the Zeeman frequency defined by the field B_0 . The behavior of the spin polarization (5) is shown in Fig. 1, where we compare the spin motion in the presence of the resonator ($h \neq 0$) and in the absence of the latter ($h = 0$). Clearly, without the resonator feedback field, the spin is blocked, while in the presence of the resonator, it reverses in short time $t_{rev} \sim 10^{-10} \text{ s}$.

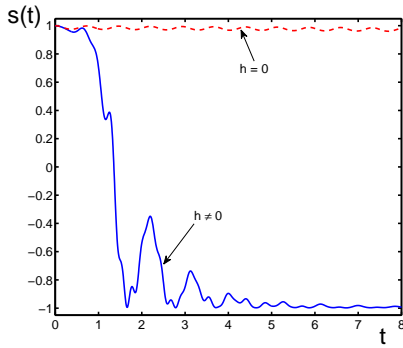


Figure 1: Spin reversal of a single nanocluster, with parameters typical of nanoclusters made of Fe, Ni, and Co.

5 Dynamics of nanocluster assemblies

The ensemble of nanoclusters is described by the Hamiltonian

$$\hat{H} = \sum_i \hat{H}_i + \frac{1}{2} \sum_{i \neq j} \hat{H}_{ij}, \quad (16)$$

where the indices $i, j = 1, 2, \dots, N$ enumerate nanoclusters. The single nanocluster Hamiltonians are

$$\hat{H}_i = -\mu_0 \mathbf{B} \cdot \mathbf{S}_i - D(S_i^z)^2 + D_2(S_i^x)^2 + D_4 [(S_i^x)^2(S_i^y)^2 + (S_i^y)^2(S_i^z)^2 + (S_i^z)^2(S_i^x)^2] , \quad (17)$$

with the total magnetic field

$$\mathbf{B} = B_0 \mathbf{e}_z + H \mathbf{e}_x . \quad (18)$$

The interaction term takes into account the dipolar spin interactions

$$\hat{H}_{ij} = \sum_{\alpha\beta} D_{ij}^{\alpha\beta} S_i^\alpha S_j^\beta , \quad (19)$$

through the dipolar tensor $D_{ij}^{\alpha\beta} = \mu_0^2 (\delta_{\alpha\beta} - 3n_{ij}^\alpha n_{ij}^\beta) / r_{ij}^3$, in which $r_{ij} \equiv |\mathbf{r}_{ij}|$, $\mathbf{n}_{ij} \equiv \mathbf{r}_{ij}/r_{ij}$, and $\mathbf{r}_{ij} \equiv \mathbf{r}_i - \mathbf{r}_j$.

One sometimes says that spin systems are similar to atomic systems, where transition dipoles are correlated by means of the photon exchange through the common radiation field. This correlation leads to coherent atomic radiation called the Dicke superradiance [34]. One says that moving spins also radiate electromagnetic field that could yield the correlated spin motion, in the same way as in the Dicke effect. To check whether this is so, we need to compare the time t_{rad} , required for inducing spin correlations through the common radiation field with the spin dephasing time T_2 . As the radiation time [35,36] for nanoclusters, we have

$$t_{rad} = \frac{3c^3}{2\hbar\gamma_S^2\omega^3 S} , \quad (20)$$

while the spin dephasing time is

$$T_2 = \frac{1}{\hbar\rho\gamma_S^2 S} . \quad (21)$$

For the typical nanocluster density $\rho \sim 10^{20} \text{ cm}^{-3}$ and $S \sim 10^3$, the spin dephasing time is $T_2 \sim 10^{-10} \text{ s}$. While for the radiation time (20), with $\omega \sim 10^{11} \text{ s}^{-1}$, we have $t_{rad} \sim 10^8 \text{ s} = 10$ years. The ratio of times (20) and (21) is extremely large: $t_{rad}/T_2 = 3c^3\rho/(2\omega^3) \sim 10^{18}$. This tells us that the spin motion in no way can be correlated through electromagnetic radiation. That is, the Dicke effect has no relation to the coherent spin motion. But spins can be correlated only through the Purcell effect requiring the presence of a feedback field caused by a resonator.

The feedback rate due to the resonator is

$$\gamma_0 = \pi\eta\rho\hbar\gamma_S^2 S . \quad (22)$$

The reversal time for N correlated nanoclusters becomes

$$t_{rev} = \frac{1}{\gamma_0} = \frac{V_{coil}}{\pi\hbar\gamma_S^2 SN} = \frac{t_{rev}^1}{N} , \quad (23)$$

where t_{rev}^1 is the relaxation time (15) for a single nanocluster inside the same coil.

We solved the evolution equations for the nanocluster assemblies involving the scale separation approach [4,14] that is a generalization of the Krylov-Bogolubov [37] averaging method. Four classes of spin objects have been investigated.

(i) Polarized nuclear materials, such as propanediol $\text{C}_3\text{H}_8\text{O}_2$, butanol $\text{C}_4\text{H}_9\text{OH}$, and ammonia NH_3 , with the parameters: $S = 1/2$, $\rho = 10^{22}\text{cm}^{-3}$, $T = 0.1\text{K}$, $B_0 \sim 10^4\text{G}$, $\omega_0 \sim 10^8\text{s}^{-1}$, $\lambda \sim 10^2\text{cm}$, $T_1 \sim 10^5\text{s}$, $T_2 \sim 10^{-5}\text{s}$, $\tau \equiv 1/\gamma \sim 10^{-6}\text{s}$. Recall that in these nanomolecules the magnetization is due to polarized proton spins.

The following characteristic times are found: thermal-noise time $t_T \sim 10^{16}\text{s} \sim 10^9$ years, radiation time $t_{\text{rad}} \sim 10^{15}\text{s} \sim 10^8$ years, and reversal time $t_{\text{rev}} \sim 10^{-6}\text{s}$.

Therefore, neither the Nyquist thermal noise nor the photon exchange through the radiated field play any role in the relaxation process. Spin dynamics, resulting in the magnetization reversal, is completely due to the action of the resonator feedback field. As is explained above, the same concerns nanomolecules and nanoclusters

(ii) Nuclear polarized ferromagnets, where proton spins are polarized and interact through hyperfine forces with electrons participating in forming ferromagnetic order. In such materials, the electron subsystem plays the role of an additional resonator enhancing effective nuclear correlations. Being interested in the motion of nuclear spins, under a fixed mean electron magnetization, we find the reversal time $t_{\text{rev}} \sim 10^{-9}$ s.

(iii) Molecular magnets, such as Mn_{12} and Fe_8 , with the typical parameters: $S = 10$, $\rho = 10^{20} - 10^{21}\text{cm}^{-3}$, $T_B = 1\text{K}$, $B_0 \sim 10^5\text{G}$, $\omega_0 \sim 10^{13}\text{s}^{-1}$, $\lambda \sim 10^{-2}\text{cm}$, $\omega_A \equiv E_A/\hbar \sim 10^{10} - 10^{12}\text{s}^{-1}$, $T_1 \sim 10^5 - 10^7\text{s}$, $T_2 \sim 10^{-10}\text{s}$. The reversal time is $t_{\text{rev}} \sim 10^{-11}$ s.

(iv) Magnetic nanoclusters composed of Fe, Ni, and Co, at $T = 1\text{K}$, with the typical parameters: $S = 10^3$, $\rho = 10^{20}\text{cm}^{-3}$, $T_B = 10 - 40\text{K}$, $B_0 \sim 10^4\text{G} = 1\text{T}$, $\omega_0 \sim 10^{11}\text{s}^{-1}$, $\lambda \sim 1\text{cm}$, $T_1 \sim 10^{34}\text{s} \sim 10^{27}\text{years}$, $T_2 \sim 10^{-10}\text{s}$. The reversal time can be very small reaching the value $t_{\text{rev}} \sim 10^{-12}$ s.

In the case of magnetic molecules and, especially, nanoclusters, because of their high spins, the system of many clusters can produce quite strong coherent radiation of the maximal intensity

$$I_{\text{max}} \sim \frac{2\mu_0^2}{3c^3} S^2 \omega^4 N_{\text{coh}}^2, \quad (24)$$

where $N_{\text{coh}} \sim \rho\lambda^3$ is the number of clusters in a coherent packet. The intensity of radiation of magnetic molecules, with $N_{\text{coh}} \sim 10^{14}$, is of order $I_{\text{max}} \sim 10^5\text{W}$. And for magnetic nanoclusters, with $N_{\text{coh}} \sim 10^{20}$, the radiation intensity can reach $I_{\text{max}} \sim 10^{12}\text{W}$.

There can happen several regimes of spin dynamics depending on the initial spin polarization, the strength of a triggering pulse, and the effective coupling parameter

$$g \equiv \frac{\gamma\gamma_0\omega_0}{\gamma_2(\gamma_2^2 + \Delta^2)}, \quad (25)$$

where $\gamma_2 \equiv 1/T_2$ and $\Delta \equiv \omega - \omega_0$ is the detuning from resonance. These regimes for nanoclusters can be classified analogously to those occurring for nuclear magnets [4,16]: incoherent free relaxation, weakly coherent free induction, weakly coherent superradiance, strongly coherent pure superradiance, strongly coherent triggered superradiance, pulsing superradiance, and punctuated superradiance [38].

It is important to stress that the existence of magnetic anisotropy in magnetic nanoclusters does not preclude the realization of fast spin reversal, provided the external magnetic field is sufficiently strong. The influence of the anisotropy energy E_A on the spin reversal of a nanocluster system is shown in Fig. 2, where $A \equiv E_A/\omega_0$. This regime corresponds to pure spin superradiance.

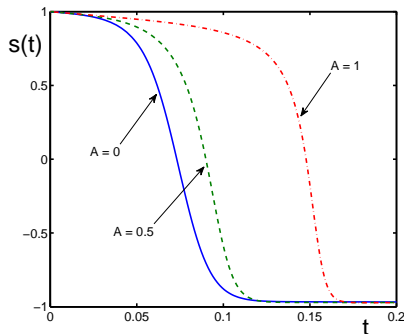


Figure 2: Influence of magnetic anisotropy on the spin reversal of an ensemble of many nanoclusters, with the parameters typical of Fe, Ni, and Co.

Coherent dynamics in the spin assemblies, formed by magnetic nanomolecules, have an important difference from the spin dynamics in an ensemble of magnetic nanoclusters. Magnetic molecules are identical and form the systems with well organized crystalline lattices. While magnetic nanoclusters vary in their shapes, sizes, and total spins, which results in an essential nonuniform broadening. Computer simulations, accomplished together with V.K. Henner and P.V. Kharebov, demonstrate that this nonuniformity does not destroy coherent spin motion. A detailed analysis of the computer simulations, with nonuniform nanocluster distributions, will be presented in a separate publication.

In conclusion, we have considered spin dynamics in magnetic nanomolecules and nanoclusters, starting from a strongly nonequilibrium state, with the magnetization directed opposite to the applied external magnetic field. We have compared several methods of describing the spin dynamics, showing that a microscopic approach, based on the quantum equations of motion, is the most accurate. We also have analyzed the influence of different effects on spin dynamics. The effects of the Nyquist-noise triggering and of Dicke correlations are found to be negligible for spin systems. This principally distinguishes spin systems from atomic systems or quantum dot systems [39], where correlations, leading to coherent radiance, are caused by the Dicke effect of interactions through the common radiation field.

The feedback field, developing in the resonator, reaches rather high values, of the order of the applied constant magnetic field. Such a strong feedback field suppresses the influence of mutual cluster interactions. Generally, in an ensemble of nanoclusters of sufficiently high density, in addition to dipole interactions, there can appear exchange interactions [40] that can influence equilibrium properties of nanoclusters. But in the considered case of strongly nonequilibrium spin dynamics, the exchange interactions are also suppressed by the self-organized resonator feedback field.

This important conclusion can be formulated as follows: *Coherent spin dynamics are completely governed by the Purcell effect that is caused by the action of the resonator feedback field.*

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